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THE NATURE OF INDIVIDUAL RADIOACTIVE PARTICLES. V. FALLOUT PARTICLES FROM SHOTS ZUNI AND TEWA, OPERATION REDWING.

I ical Report USNRDL-TR-133

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by

C.E. Adams

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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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THE NATURE OF INDIVIDUAL RADIOACTIVE PARTICLES. V. FALLOUT PARTICLES FROM SHOTS ZUNI AND TEWA, OPERATION REDWING.

Research and Development Technical Report USNRDL-TR-133

NS 081-001
and U.S. Army

1 February 1957

by

C.E. Adams

Military Applications

Technical Objective
AW-7

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ABSTRACT

Studies were made of the structure and composition of, and the distribution of radioactivity within,the fallout particles resulting from Shots Zuni and Tewa, Operation REDWING. Techniques utilizing X-ray diffraction analysis and the petrographic microscope were employed. It was found that most of the radioactive fallout particles were composed of calcium hydroxide, or a mixture of calcium hydroxide and calcium oxide, with a surface layer of calcium carbonate. The particles were formed by the hydration and carbonation of calcium oxide which had been formed by the heat of the bombs acting on the coral (calcium carbonate) of the islands at the site. The radioactivity was found on the surface of those particles which had not been heated sufficiently to melt and was usually found throughout the volume of those particles which had melted.

This is the fifth in a series of reports on the nature of fallout particles: part I, USNRDL-374, 28 November 1952; II, USNRDL-408, 1 July 1953; III, USNRDL-440, 24 February 1954; IV, USNRDL-TR-26, 17 January 1955; all by C.E. Adams.

SUMMARY

The Problem

This is the fifth in a series of reports devoted to the study of radioactive fallout particles originating in various types of physical environment. The fallout particles described here were collected following Shots Zuni and Tewa, Operation REDWING. Both shots were surface bursts over coral islands.

The chemical composition of the particles and the structure and the distribution of the chemical compounds within the particles were determined by studying thin sections of the particles under the petrographic microscope. Determinations of the chemical composition were also made by X-ray diffraction analysis.

Findings

In general, two shapes of particles were found. The first group was angular and composed of calcium hydroxide throughout with a surface layer of calcium carbonate. The radioactivity occurred on the surfaces of these particles. The second group were spheroidal and were composed of mixtures of calcium oxide and calcium hydroxide with a surface layer of calcium carbonate. The radioactivity was usually found distributed throughout the volumes of these particles.

The fallout particles were formed through the decarbonation of the calcium carbonate coral grains by the heat of the bomb and through the subsequent hydration and carbonation of the calcium oxide. The spheroidal particles had been heated sufficiently to melt the calcium oxide. The radioactivity collected on the surfaces of the particles. In the case of the liquid drops, the radioactivity mixed throughout the volume of the particles.

ADMINISTRATIVE INFORMATION

This work was sponsored by the Bureau of Ships. The Program is described in U.S. Naval Radiological Defense Laboratory Annual Report to the Bureau of Ships (DD Form 613), NS 081-001, Subtask 1, paragraph 4, of July 1956 (Encl (1) to CO and Dir, USNRDL Secr ltr 3-905-471 EHC:dlc, Ser 0014921 of 31 August 1956).

The fallout particles were collected at Operation REDWING as part of Project 2.6.3 as described in U.S. Naval Radiological Defense Laboratory Annual Report to the Bureau of Ships (DD Form 613) for Project NS 088-001, Subtask 4B, of February 1956 (Encl (1) to CO and Dir, USNRDL Secr ltr 3-905-335 Ser 0014173 of 16 March 1956.

The work also is part of the technical program for the Department of the Army established between Department of the Army, Office, Chief of Research and Development, and Bureau of Ships (Joint Agreement, 23 November 1955).

This report is the fifth in a series on the nature of fallout particles: Part 1, USNRDL-374, 28 November 1952; II, USNRDL-408; 1 July 1953; III, USNRDL-440, 24 February 1954; IV, USNRDL-TR-26, 17 January 1955; all reports are by C.E. Adams.

INTRODUCTION

A study has been made of the chemical composition, structure and distribution of radioactivity in the radioactive fallout particles resulting from Shots Zuni and Tewa, Operation REDWING at the Eniwetok Proving Grounds. Both shots involved large-yield weapons. Zuni was detonated on the surface of an island and Tewa was detonated on a barge anchored in shallow water.

The radioactive particles were selected from fallout samples collected on the deck of a barge, the YFNB-29, which was anchored in Bikini lagoon a few miles from the shot point and used as a floating platform for fallout collectors and other instruments. These particles represented close-in fallout and were relatively large, ranging in size from about 1/2 to 2 mm in diameter. The particles were mounted in small plastic pellets which were subsequently ground into thin sections using standard mineralogical techniques. The thin sections were cut through the center of each particle and were about 30 μ thick. By studying these thin sections under the petrographic microscope it was possible to determine the composition and structure of the fallout particles. The composition of the particles was checked further by X-ray diffraction analysis.

The distribution of radioactivity within the particles was found by radioautographing the thin sections with Eastman NTB stripping film.

PARTICLE CHARACTERISTICS

Two general types of particles were observed. The first type, greater in number than the other, was angular in shape and a dead white in color.

Note. This report is the fifth in a series on the nature of fallout particles: part I, USNRDL-374, 28 November 1952; part II, USNRDL-408, 1 July 1953; part III, USNRDL-440, 24 February 1954; part IV, USNRDL-TR-26, 17 January 1955; all reports are by C. E. Adams.

These particles resembled coral sand grains in appearance except for the unusual purity of the white color.

The second type was spheroidal and white to pale yellow. Many of these particles contained a central cavity which often opened through the side of the particle.

Angular Particles

The angular particles were composed of fine-grained calcium hydroxide with a thin outer layer of calcium carbonate (Fig. 1). Occasionally, a particle was found that contained a central core of unaltered coral (calcium carbonate) surrounded by an outer zone of calcium hydroxide. Rarely, a radioactive particle was found that was composed wholly of unaltered coral (Fig. 2).

The radioactivity associated with the angular type particles was always found on the surfaces of the particles. In the case of particles originating at Shot Tewa, the radioactivity was distributed more or less evenly over the surfaces but with some spots and areas of higher intensity. Apparently, the activity had diffused a short distance into each particle. Examination of the radioactive areas of the thin sections with high magnification failed to reveal any tiny particles adhering to the surfaces of the angular particles which might have been the ultimate carriers of the radioactivity. However, when some of the particles were powdered and examined with an electron microscope, a few spheres on the order of a few tenths of a micron in diameter were observed.

The occurrence of the radioactivity on the angular fallout particles from Shot Zuni was similar to that on the Tewa particles. Examination under high magnification of the areas of the particles covered with the even, homogeneous distribution of radioactivity failed to reveal any small particles as carriers of the radioactivity. However, associated with many of the spots and areas of more intense radioactivity were small particles adhering to the surfaces of the large angular particles. These small particles were spherical in shape and varied in size from about 1 to 25 μ in diameter, most of them being in the range 2 to 10 μ in diameter (Fig. 3). The smallest of these particles were transparent and were orange-red in color. The larger ones were opaque and appeared black by reflected light.

While these particles were so small that it was impossible to determine their composition directly, their composition might be guessed by comparing them with similar-appearing fallout particles collected following a low-yield tower shot, Inca, Operation REDWING. The Inca particles were of the same shape and color as these particles but were many times larger so that it was possible to determine their composition by X-ray diffraction analysis. They were composed mostly of Fe_3O_4 and $2CaO.Fe_2O_3$ with smaller amounts of Fe_2O_3 and CaO. Many of the particles were pure iron oxide but most contained some calcium oxide either free or combined with the iron oxide as calcium ferrite. These particles originated from the condensation of the vaporized iron from the tower and other structures and from vaporized coral soil.*

Spheroidal Particles

The spheroidal fallout particles were composed of coarse-grained calcium hydroxide with inclusions of calcium oxide and with a surface layer of calcium carbonate (Fig. 4). In some cases the calcium oxide made up the greater part of the volume of the particles (Figs. 5 and 6) but in most of the particles the oxide had been largely converted to the hydroxide and such oxide as remained was found disseminated in small grains throughout the particle.

The radioactivity associated with these spheroidal particles was usually found throughout the volume of the particle occurring both as diffuse areas and as single hot spots. On some of the spheroidal particles the radioactivity was confined near the surface as in the case of the angular particles. Under high magnification there were no observable sources or small particles carrying the radioactivity even in the cases where the activity was observed to occur in spots.

ORIGIN OF PARTICLES

The fallout particles originated through the action of the heat and the vaporized fission products from the bomb on the coral sand of the atoll islands. The coral sand is composed chiefly of calcium carbonate which decomposes to calcium oxide and carbon dioxide at about 800 to 900°C. Calcium oxide melts at 2570°C. As previously described, the majority of fallout particles consist of angular grains of calcium hydroxide with radioactivity deposited on their surfaces. These particles were formed by the heating of the coral sand grains, which had been thrown aloft by the force of the explosion and the associated atmospheric turbulence, sufficiently to convert them to calcium oxide but not sufficiently to melt the calcium oxide. These particles still retain the angular shape and microcrystalline structure of the original coral sand grains.

* A detailed report on these particles is to be published as part VI in this series.

Much of the radioactivity on the angular Zuni particles was due to the collection by impaction of small radioactive spheres. As described above, these small spheres probably originated from the condensation of vaporized metals in the bomb and associated structures, of vaporized calcium oxide, and of vaporized fission products.

The origin of the radioactivity on the Tewa particles and on those Zuni particles which do not carry any visible small particles is not known. The radioactivity could have collected on these particles by the impaction of tiny radioactive particles which were either too small to be visible in the optical microscope or whose optical characteristics were too similar to those of the large fallout particles to permit their differentiation; or the radioactivity could have collected by the direct condensation of vaporized fission products, or fission-product oxides, on the surfaces of the large calcium oxide particles.

The fact that a few submicroscopic spheres were found among the powdered Tewa particles indicates that at least some of the radioactivity was accumulated by the impaction of previously condensed particles. On the other hand, the concentrations of the vaporized metals and metallic oxides, both fission products and structural materials, were sufficiently low in the fireball so that even those constituents with low vapor pressures can remain stable in the vapor phase at fireball temperatures down below the melting point of calcium oxide. This indicates that the solid calcium oxide particles could have served as a condensation surface for the vaporized fission products.

Either following or, perhaps, concurrently with the initial heating and decarbonation of the particles, the calcium oxide was hydrated to calcium hydroxide probably by the water vapor produced from the sea water by the heat of the explosion. During this process a portion of the surface radioactivity apparently was soluble enough so that it diffused slightly into the particle. Finally, the surface layer of the calcium hydroxide was converted to calcium carbonate by carbon dioxide in the atmosphere.

While most of these particles were heated sufficiently to decarbonate the coral, several radioactive particles were found which consisted of unaltered coral grains or coral grains which had been heated only enough to decarbonate an outer layer. The radioactivity on these particles occurred both as a homogeneous deposit over the surface and as spots associated with small, highly active particles adhering to the surfaces of the coral grains.

Many other unaltered coral grains which were inactive were found in the collections from the YFNB-29.

Rarely, a particle was found that consisted of partially decarbonated coral but which was nonradioactive, indicating that it was possible for soil material to have been exposed to moderate heat in the cloud but to have suffered no radioactive contamination.

The origin of the spheroidal fallout particles was similar to that of the angular particles except that they were heated sufficiently to melt the calcium oxide and form liquid drops. In many cases, the heating was apparently quite rapid so that the exterior of the particle had already melted before all the carbon dioxide resulting from the decarbonation of the original coral had had time to escape from the interior of the particle. This caused many of the particles to have hollow spaces in their centers.

There is the possibility that these large spheroidal particles could have been formed by a series of collisions of smaller liquid drops. The fact that the radioactivity was often found scattered irregularly throughout the large spheres would indicate this. However, the fact that many of the large spheroids have radioactivity only on or near their surfaces and that many of them are hollow would be arguments against their formation by an accretion process but that they were formed, as were the angular particles, from individual coral sand grains.

As was the case with the angular particles, the radioactivity could have been collected either by the impaction of previously condensed small radioactive spheres or by the direct condensation of vaporized fission products on the liquid calcium oxide drops. In either case, the radioactivity was probably partially dissolved and distributed throughout the volume of the liquid particle by diffusion and convection.

In contrast to the angular fallout particles, most of the spheroidal particles were incompletely hydrated and still retained about 5 to 75 per cent by volume unaltered calcium oxide. The complete hydration of the angular particles was probably due to the formation of porous calcium oxide upon decarbonation of the original calcium carbonate particles. This porous structure would be quite reactive with water vapor and would be easily and rapidly hydrated. On the other hand, the spheroidal particles formed by the solidification of fused calcium oxide would be dense and nonporous and reaction with water vapor would proceed much more slowly.

In addition to the categories of angular and spherical particles there were observed particles shaped like fluffy aggregates. As these particles were too fragile to be made into thin sections nothing is known of their structure. However, several of them were crushed on a microscopic slide and examined under the petrographic microscope. They were found to be composed mostly of calcium hydroxide with some calcium carbonate and there was nothing to indicate that they differed significantly from the other types of particles.

There occurs a 98-per cent increase in volume when calcium oxide hydrates to calcium hydroxide and it has been observed that small pellets of calcium oxide placed in distilled water will expand into fragile, fluffy structures similar to those observed among the fallout particles. It seems probable that the fluffy fallout particles have originated in some similar manner wherein calcium oxide particles have come into contact with drops of water or a fine mist in the air. It has also been observed that many of the solid fallout particles change into fluffy aggregates or grow fluffy appendages after standing for several weeks in air.

While the samples from which the thin sections were made were collected from one locality only, it is nevertheless believed that the particles described are representative of the total radioactive fallout material. Samples of radioactive fallout particles collected at distances of approximately forty to eighty miles from the shot point on the converted Liberty Ships (YAG-39 and YAG-40) used by this laboratory were identical in appearance, except for being smaller in size, to those collected on the barge, YFNB-29.

Also, the particles described in this report are exactly the same in composition and structure as the radioactive fallout particles collected over wide areas following shots similar to Zuni and Tewa at Operations IVY[1] and CASTLE[2].

Approved by:
L. B. Werner

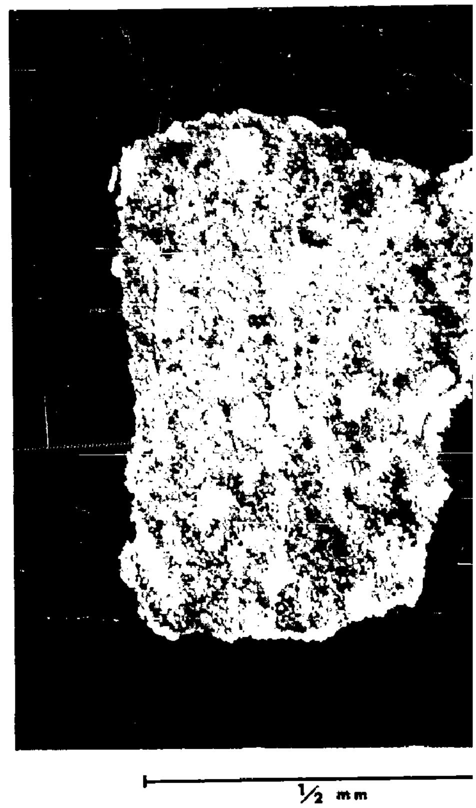
for E.R. TOMPKINS
Head, Chemical Technology Division

For the Scientific Director

1a

1c

2a



2c

Fig. 2 An Angular Fallout Particle
Zuni. This particle is unusual in th
posed wholly of unaltered coral.
2a. Ordinary Light. The structure
is visible.
2b. Crossed Nicols. The particle i
composed of calcium carbonate thr
(photographs white).
2c. Radioautograph. The radioact
mostly as spots on the surface and in
is associated with small, black sphe
to the surface of the particle. The
seen by careful examination of the
thin section in Fig. 2a which corres
the most active areas as shown in th
graph.

Fig. 3 Photograph Under High Magnification of Part of an Angular Fallout Particle From Shot Zuni. Observe the large numbers of small, black spheres adhering to the surface. These spheres are carriers for much of the radioactivity on many of the angular Zuni particles but were not observed on the Tewa angular particles nor on any of the spheroidal particles.

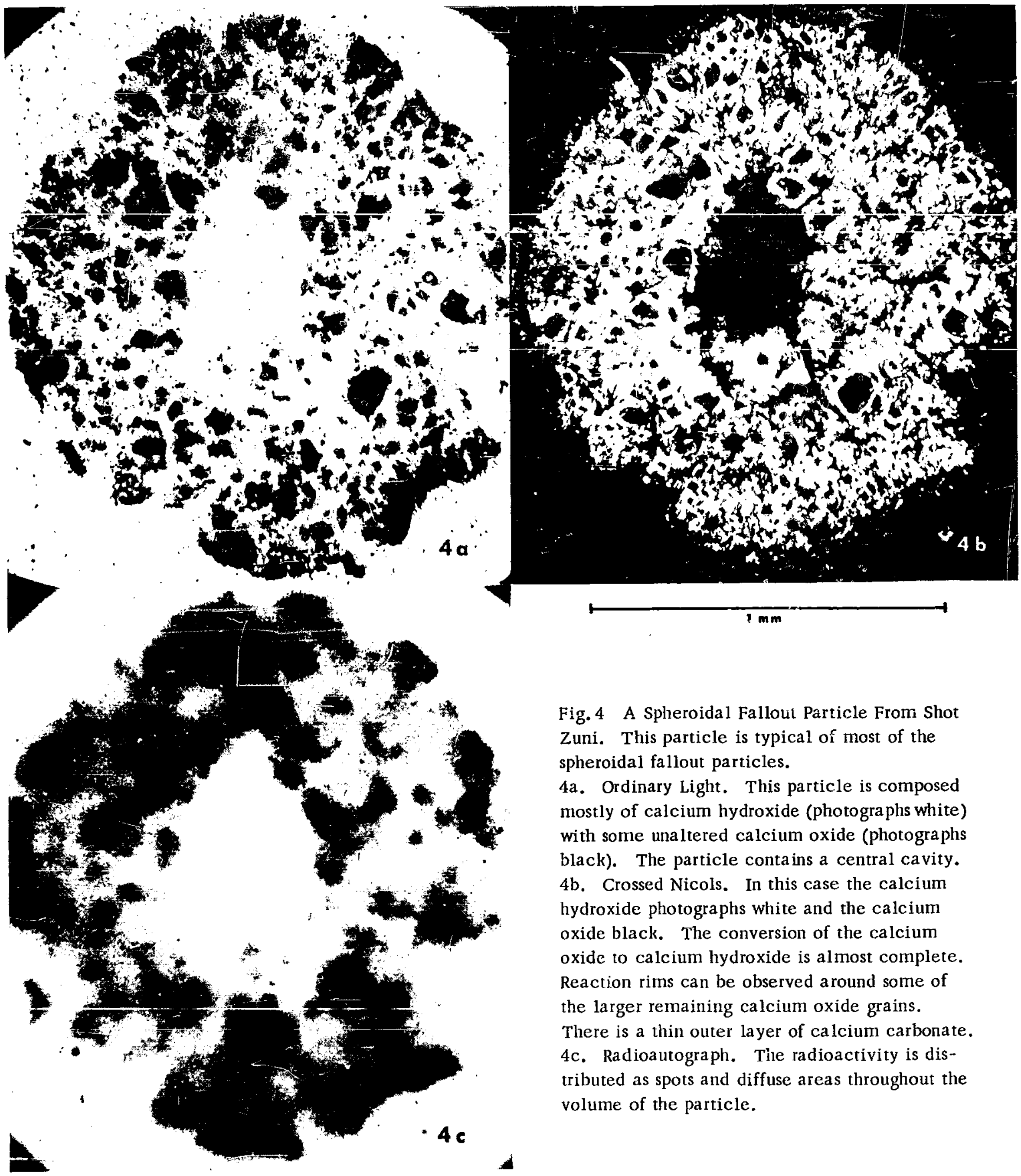


Fig. 4 A Spheroidal Fallout Particle From Shot Zuni. This particle is typical of most of the spheroidal fallout particles.
4a. Ordinary Light. This particle is composed mostly of calcium hydroxide (photographs white) with some unaltered calcium oxide (photographs black). The particle contains a central cavity.
4b. Crossed Nicols. In this case the calcium hydroxide photographs white and the calcium oxide black. The conversion of the calcium oxide to calcium hydroxide is almost complete. Reaction rims can be observed around some of the larger remaining calcium oxide grains. There is a thin outer layer of calcium carbonate.
4c. Radioautograph. The radioactivity is distributed as spots and diffuse areas throughout the volume of the particle.

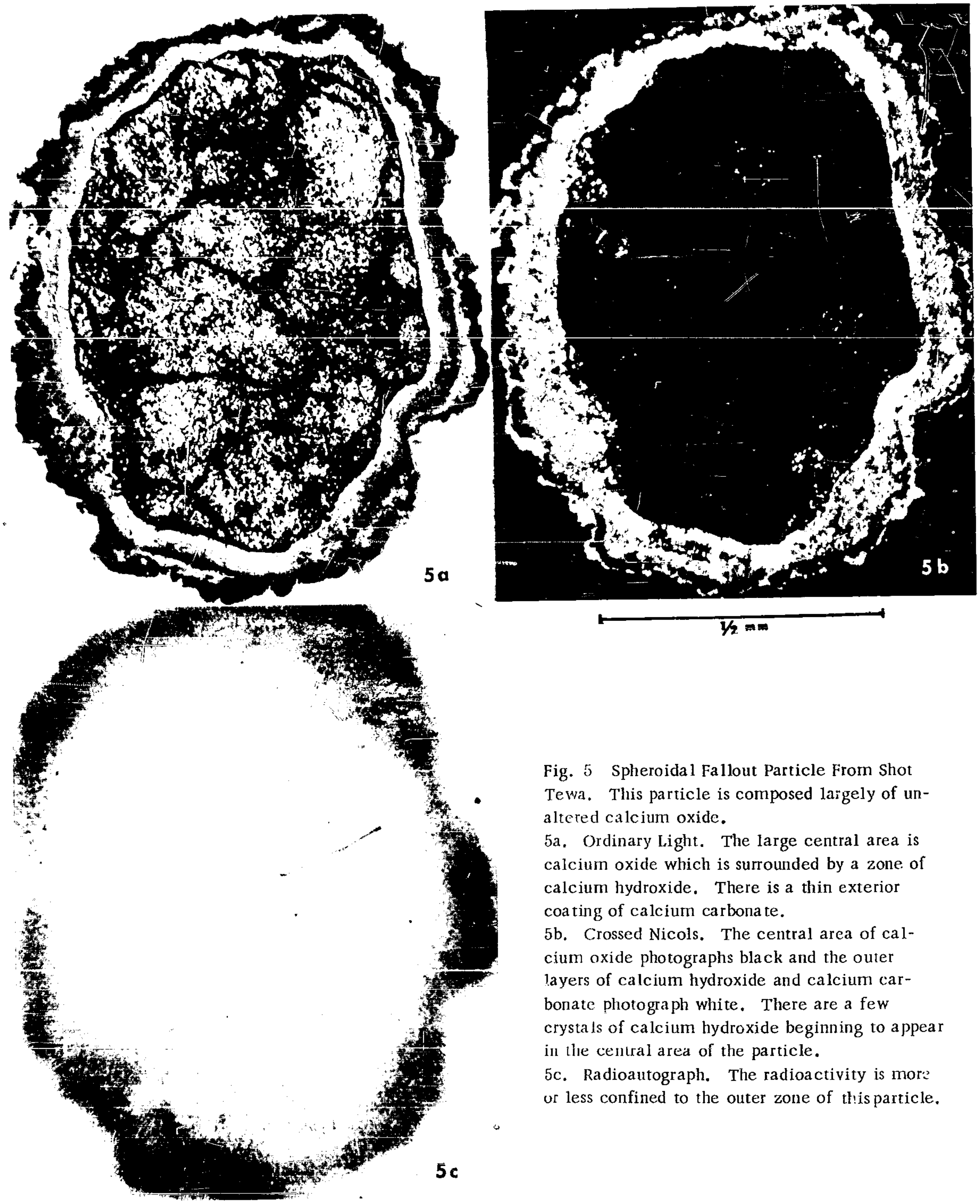


Fig. 5 Spheroidal Fallout Particle From Shot Tewa. This particle is composed largely of unaltered calcium oxide.

5a. Ordinary Light. The large central area is calcium oxide which is surrounded by a zone of calcium hydroxide. There is a thin exterior coating of calcium carbonate.

5b. Crossed Nicols. The central area of calcium oxide photographs black and the outer layers of calcium hydroxide and calcium carbonate photograph white. There are a few crystals of calcium hydroxide beginning to appear in the central area of the particle.

5c. Radioautograph. The radioactivity is more or less confined to the outer zone of this particle.

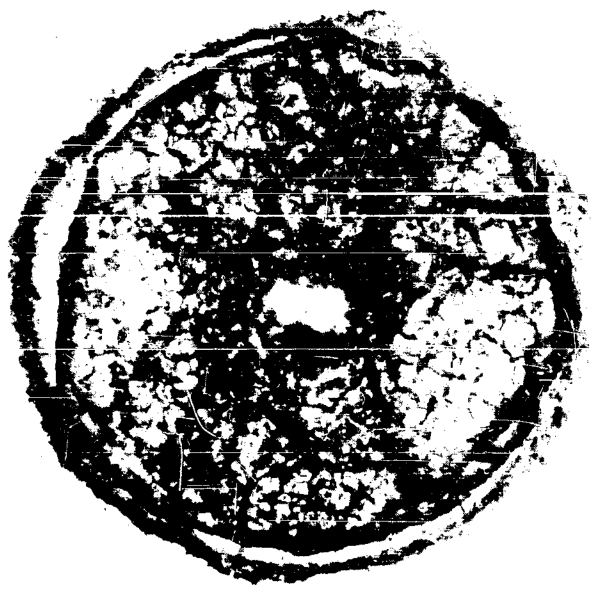


Fig. 6 A Spheroidal Particle From Shot Zuni. This particle consists mostly of unaltered calcium oxide with outer layers of calcium hydroxide and calcium carbonate and is similar to the particle shown in Fig. 5 except that the radioactivity is distributed throughout the volume of the particle.

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This is the fifth in a series of reports on the nature of fallout particles: part I, USNRDL-374, 28 November 1952; II, USNRDL-408, 1 July 1953; III, USNRDL-440, 24 February 1954; IV, USNRDL-TR-26, 17 January 1955; all by C.E. Adams.

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Studies were made of the structure and composition of, and the distribution of radioactivity within, the fallout particles resulting from Shots Zuni and Tewa, Operation REDWING. Techniques utilizing X-ray diffraction analysis and the petrographic microscope were employed. It was

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found that most of the radioactive fallout particles were composed of calcium hydroxide, or a mixture of calcium hydroxide and calcium oxide, with a surface layer of calcium carbonate. The particles were formed by the hydration and carbonation of calcium oxide which had been formed by the heat of the bombs acting on the coral (calcium carbonate) of the islands at the site. The radioactivity was found on the surface of those particles which had not been heated sufficiently to melt and was usually found throughout the volume of those particles which had melted.

This is the fifth in a series of reports on the nature of fallout particles: part I, USNRDL-374, 28 November 1952; II, USNRDL-408; 1 July 1953; III, USNRDL-440, 24 February 1954; IV, USNRDL-TR-26, 17 January 1955; all by C.E. Adams.

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Defense Threat Reduction Agency
45045 Aviation Drive
Dulles, VA 20166-7517

CPWS/TRC March 26,1999

MEMORANDUM TO DEFENSE TECHNICAL INFORMATION CENTER
ATTN: OCQ: MR WILLIAM BUSH

SUBJECT: DOCUMENT REVIEW

The Defense Threat Reduction Agency's Security Office has reviewed and declassified the following documents:

USNRDL-TR-133, **AD-145694**, UNCLASSIFIED, STATEMENT A
URS-B162-6, **AD-349217**, UNCLASSIFIED, STATEMENT D, ADMINISTRATIVE OR OPERATIONAL USE, 3/19/99
USNRDL-TR-215, **AD-C006187**, UNCLASSIFIED, STATEMENT A
URS-162-8, **AD-348723**, UNCLASSIFIED, STATEMENT D, ADMINISTRATIVE OR OPERATIONAL USE, 3/19/99
WES-TR-2-471, **AD-909045**, UNCLASSIFIED, STATEMENT C, TEST AND EVALUATION, 3/9/72.
WES-TR-1-695, **AD-368244**, UNCLASSIFIED, STATEMENT A
WES-MP-1-689, **AD-356460**, UNCLASSIFIED, STATEMENT C, ADMINISTRATIVE OR OPERATIONAL USE, 3/23/99

These documents were reviewed under the Executive Order 12958.

Ardith Jarrett

ARDITH JARRETT
Chief, Technical Resource Center

Completed
7 Jun 2000
B.W.